LABORATORY Spotlight
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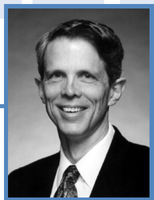
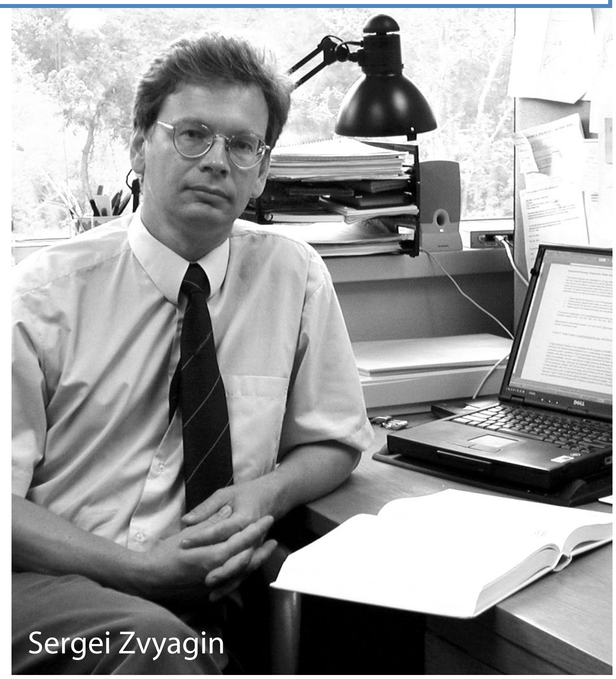


Attention Users

Bruce Brandt
Director, NHMFL DC Field User Program

Sergei Zvyagin and Jurek Krzystek have recently completed a new facility for spectroscopy between 140 and 700 GHz in a 25 T high homogeneity resistive magnet in the DC High Field Facility of the NHMFL in Tallahassee. This new equipment is complete and has already attracted several users. It is described in detail in the article below.



Sergei Zvyagin

Features of and First Results from a New Submillimeter/Millimeter Wave Spectroscopy Facility at the DC Facility in Tallahassee

Sergei Zvyagin, NHMFL/FSU
Jurek Krzystek, NHMFL/FSU

Millimeter and submillimeter (hereafter combined as submillimeter/millimeter) wave spectroscopy plays an important role connecting far-infrared and conventional microwave[1] spectral methods. It covers the frequency range of 30 to 3000 GHz, which spans the energy, temperature, and magnetic field scales relevant to numerous fascinating phenomena in condensed matter science, chemistry, and biology. It is an extremely powerful tool to study magnetic excitation spectra in solids, liquids, and gases, providing very valuable information concerning magnetic structure and interactions in many substances.

Traditional (or multi-frequency) electron-spin resonance (ESR) spectroscopy employs one constant frequency (or a set of frequencies). The spectrometer that we recently built at the NHMFL's DC High Field Facility in Tallahassee, allows for operation in a very wide, quasi-continuously covered, frequency range of 140 to 700 GHz (wavelength range of ~2.1 to 0.43 mm). This range can be expanded by buying more sources. The frequency resolution is remarkable—better than 0.05 GHz *in the entire frequency range*! A key feature of the spectrometer is the set of easily-tunable, highly-monochromatic, stable, relatively powerful microwave sources, *Backward Wave Oscillators* (BWOs). These radiation sources in combination with the highly-homogeneous (better than 10^{-5}/cm) magnetic field provided by a 25 T resistive magnet (built with financial assistance from the W.M. Keck Foundation) makes the facility in Tallahassee *unique* in solving a large number of scientific problems in this frequency-field range. Some examples of measurements that may be done include the study of elementary excitation spectra in highly-correlated electron systems; spin dynamics in quantum low-dimensional and spin-ordered materials; single-molecule magnetism; electron and magnetic structure of solids; ferromagnetic, antiferromagnetic, and cyclotron resonance phenomena; physics of field-induced and spontaneous phase transitions; high-resolution ESR spectroscopy of transition metal ions (which is of great importance in chemistry, biochemistry, and structural biology); and ESR on paramagnetic ions with large zero-field splitting.

Backward wave oscillators appeared as a result of intensive work by scientists and engineers shortly after

World War II. The development of microwave electronics towards the short-wavelength part of the electromagnetic spectrum was extremely important for improvements to radar and high bandwidth communications. A set of short-length BWOs was developed and brought to industrial production in the 1960s in France and the U.S.S.R. In the 1970s, solid-state generators (based on IMPATT and Gunn diodes) began to replace BWOs for most applications because of their lighter weight, smaller size, and so forth. Industrial production of short-wave BWOs was significantly reduced, and seems to have survived only in Russia.

A BWO (Fig. 1) is a classic vacuum-tube microwave device similar to a klystron, magnetron, or a traveling wave oscillator. It possesses an important distinguishing characteristic: it is tunable over a very wide frequency range—up to ±30% from its central frequency. The main part of a BWO is a corrugated comb-like electrode called a slowing system. Interaction of the electron beam and the variable potential of the slowing system results in velocity/phase modulation of the electrons. The periodically grouped electron bunches continue to interact with the variable potential, producing an electromagnetic wave traveling in the opposite direction (backward wave). The velocity of the electrons and, thus, the radiation frequency are determined by the magnitude of the accelerating field. The BWO needs to be adjusted in the high magnetic field by rotating it around two axes. BWO is highly sophisticated device, working in an extremely intensive mode (at high voltage - up to 5-6 kV, high temperature of cathode—up to 1200°C, with high electron beam current density—up to 150 A/cm^2).

The present configuration of the submillimeter/millimeter spectrometer includes a set of four BWOs,

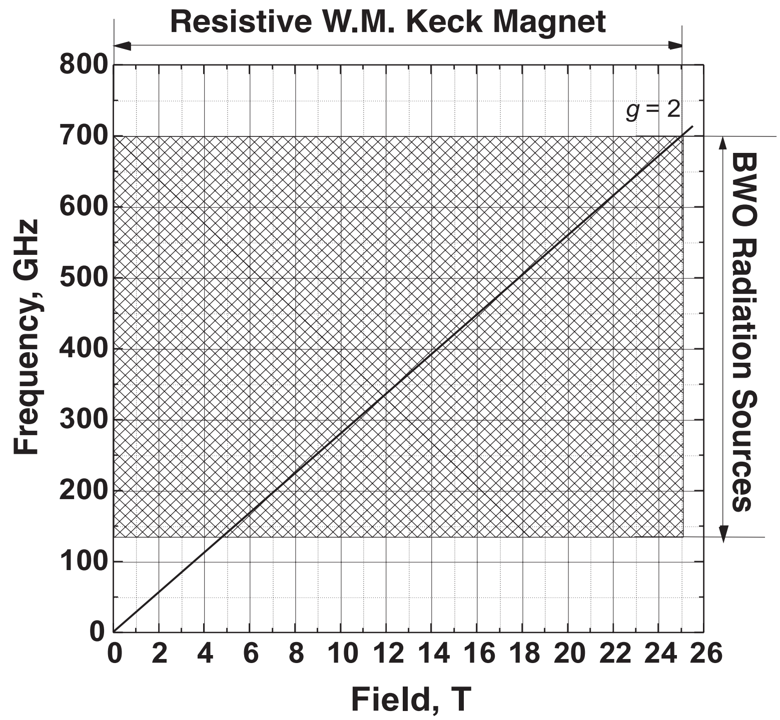


Figure 2. The frequency-field range of the facility. The line shows the frequency-field dependence of excitations with g = 2.

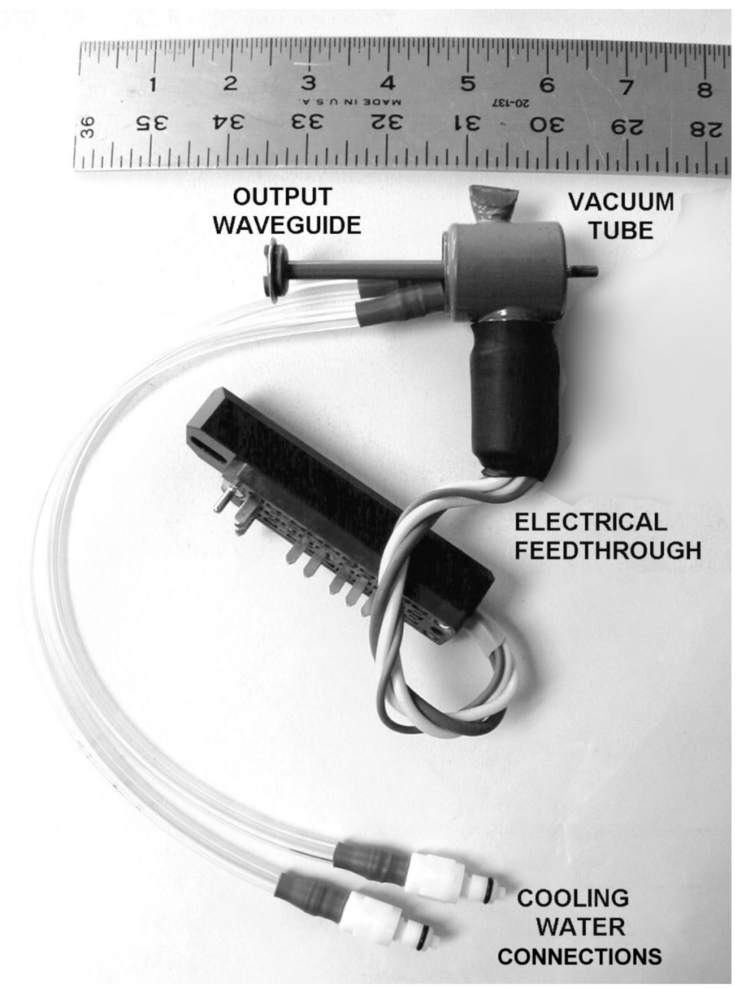


Figure 1. A backward wave oscillator.

covering the frequency ranges of 140 to 260, 200 to 380, 320 to 550, and 450 to 700 GHz with average outputs of 80, 25, 10 and 5 mW, respectively. The output power is a rather complex function of the anode voltage. The long-term frequency stability of the BWOs is better than $\Delta f/f \sim 10^{-5}$. The spectrometer works in transmission mode and employs oversized cylindrical waveguides. The spectrometer allows for experiments at sample temperatures from 1.6 to 300 K. An extremely low-noise, wide frequency range, InSb hot electron bolometer, operated at liquid-He temperature, serves as a detector. The spectra are recorded during the magnetic field sweeping. Two kinds of signal modulation are possible. While modulation of the magnetic field gives a better signal-to-noise ratio for narrower lines, modulation of the microwave power (optical modulation) allows one a direct detection of the absorption/transmission and provides better sensitivity for broader resonance lines. The spectrometer operates in the Voigt or Faraday geometry (magnetic component of the radiation parallel or perpendicular to the external magnetic field, respectively).

The facility was used to map field-induced structural phase transitions and to study resonance properties of the slightly doped spin-Peierls compound $CuGeO_3$ across the different regions of its phase diagram.[2] It is known that $CuGeO_3$, the first inorganic spin-Peierls material, undergoes a continuous structural transition from an undistorted uniform (U) phase with a gapless excitation spectrum to a gapped dimerized (D) phase with a collective nonmagnetic spin-singlet ground state at $T_{SP} \sim 14.3$ K.[3] Below T_{SP}, high magnetic field induces a transition to a new magnetic phase in which an incommensurate (I) lattice modulation is stabilized by the Zeeman energy. The field-induced lattice and spin incommensurability results in a magnetic soliton structure, where dimerized regions are separated by domain walls. It was shown that the soliton-like structure in $CuGeO_3$ exists only in a narrow range of magnetic fields close to the D-I transition.[4] This transition occurs in $CuGeO_3$ +

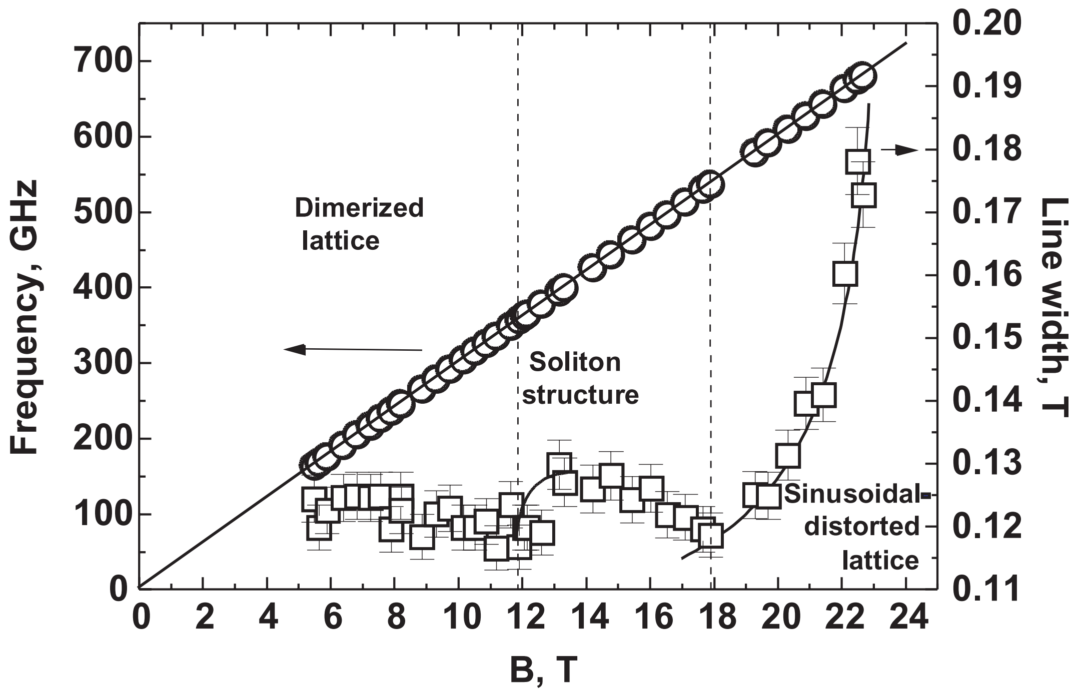


Figure 3. The ESR frequency-field dependence (circles) and the line-width, $\Delta B/B$ behavior (squares) in $CuGeO_3$ + 0.4%Si, *B // a*, T = 4.2 K. The lines are guides to eye.

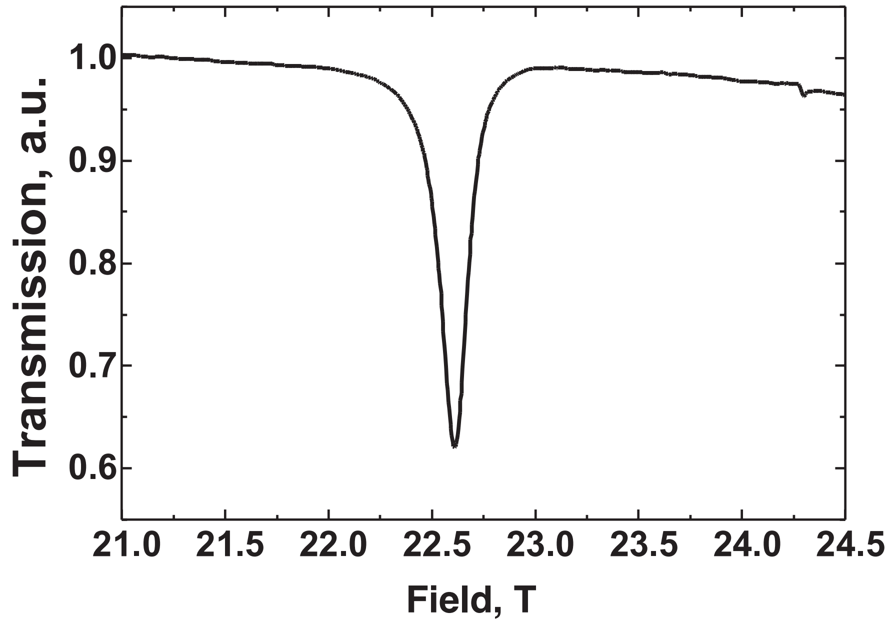


Figure 4. A high-field ESR spectrum of the spin-Peierls compound $CuGeO_3$ + 0.4%Si in the sinusoidal-distorted incommensurate phase. Frequency is 680 GHz, T=4.2 K. The small feature at ~ 24.3 T is a resonance of DPPH that is used as a marker.

0.4%Si at B ~ 12 T and can be seen as a slight change in the ESR line width in Fig. 3. Further developing of the magnetic structure, namely a sinusoidal distortion at higher fields,[5] results in a significant line broadening (more than 50 %) observed by us in the field range of 18 to 23 T. This line broadening corresponds to a continuous change from a soliton-like structure to a high-field sinusoidal-distorted phase and can be explained in terms of a sinusoidal distribution of local environments. Note that the g-factor measured along the *a*-axes does not depend on magnetic field and remains constant (g = 2.15) in the field range of ~ 5 to 23 T. This observation is consistent with results of ESR in pulsed magnetic fields.[6] A typical high-field ESR spectrum of $CuGeO_3$ + 0.4%Si in the sinusoidal-distorted incommensurate phase is shown in Fig. 4. Our results obtained in the wide frequency-field range using new submillimeter/millimeter facility offer a good possibility to test quantitatively various numerical and analytical models developed to describe field-induced structural phase transitions in spin-Peierls materials.

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1 "Microwave" is used here in the traditional sense to mean any electromagnetic wave with wavelength less than a meter.

2 The work was done in collaboration with P.H.M. van Loosdrecht (University of Groningen, The Netherlands), G. Dhalenne and A. Revcolevschi (Université de Paris-Sud, France)

3 Hase, M., *et al.*, *Phys. Rev. Lett.*, **70**, 3651 (1993).

4 Lorentz, T., *et al.*, *Phys. Rev. Lett.,* **81**, 148 (1998).

5 Ronnow, H.M., *et al.*, *Phys. Rev. Lett.*, **84**, 4469 (2000).

6 Nojiri, H., *et al.*, *Physica B*, **16**, 246-247, (1998).